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OZONE AND NO_2 MEASUREMENTS FROM ABERYSTWYTH AND LERWICK

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ABSTRACT

Measurements of the total column of ozone and NO_2 were obtained by a SAOZ UV/Visible spectrometer at Aberystwyth (52.4°N, 4.1°W) and Lerwick (60.1°N, 1.2°W) during the period March 1991 - April 1992. NO_2 measurements show a marked decrease in 1992 compared with 1991, due to the effect of aerosols from Mt. Pinatubo. Ozone measurements appear to have been affected by the aerosols - comparisons with both Dobson and TOMS measurements are presented.

1. INTRODUCTION

The SAOZ (System d'Analyse par Observation Zenitale) began operating at Aberystwyth (52.4°N, 4.1°W) on 7th March 1991. It is a UV-Visible spectrometer system, developed by CNRS in Paris (Pommereau and Goutail, 1988), which uses a diode array detector to measure the spectrum of sunlight scattered by the atmosphere at the zenith. In addition, in this later model, a pointing telescope allows direct observations of the sun and moon (although zenith sky measurements only will be presented here). The spectrometer operates over the wavelength range 300 to 600 nm. A Hewlett-Packard computer controls the acquisition of spectra, analyses the measurements and stores the spectra on floppy disks.

Software supplied by CNRS compares the spectra with a reference spectrum, obtained at high altitude with the Sun high in the sky, to determine the excess absorption at wavelengths absorbed by NO_2 (430-450 nm) and O_3 (500-560 nm). All zenith sky measurements shown here were obtained using the same version of the analysis program (version 92.426) and are restricted to solar zenith angles 87-91°. The vertical column is derived from:

$$N_V = \frac{N_S + N_R}{AMF}$$

where N_V = vertical column total of absorber
N_S = measured slant column of absorber
N_R = amount of absorber in reference spectrum
AMF = Air mass factor - determined by geometry of observation and vertical profiles of absorbers and scatterers.

At twilight, the very long path of light through the atmosphere increases the sensitivity of the measurement and reduces its susceptibility to uncertainties in the amount of absorber in the reference spectrum. Unfortunately, as noted below, the air mass factor calculation is sensitive to the vertical distribution of scatterers. Since the eruption of Mt. Pinatubo in June 1991 (and especially since the bulk of the resulting aerosol cloud appeared in Aberystwyth in early December) the conversion of slant to vertical column has been somewhat unreliable at zenith angles > 90°.

2. MEASUREMENTS AT ABERYSTWYTH: MARCH-OCTOBER 1991

The SAOZ was operated at Aberystwyth during the summer of 1991. During this period two methods were used to determine the amount of ozone in the reference spectrum:

a) If the wrong amount is used for N_R, a spurious diurnal variation is introduced into N_V, since daytime measurements are much more sensitive to N_R than those at twilight (because of the smaller AMF). Data for clear days were examined and N_R altered until the diurnal variation was removed. The optimum value was found to be 575 ± 25 Dobson Units (DU).
b) Again for clear days, Langley plots of slant column versus AMF were drawn, and the intercept calculated by linear regression. Values obtained this way were consistent with those obtained using a), but with poorer precision.

For NO_2, the value of $N_R = 8.7 \times 10^{15}$, supplied by CNRS, was retained.

3. MEASUREMENTS AT LERWICK

At the end of October 1991 the spectrometer was moved to Lerwick, Shetland Islands (60.1°N, 1.2°W), to be operated as

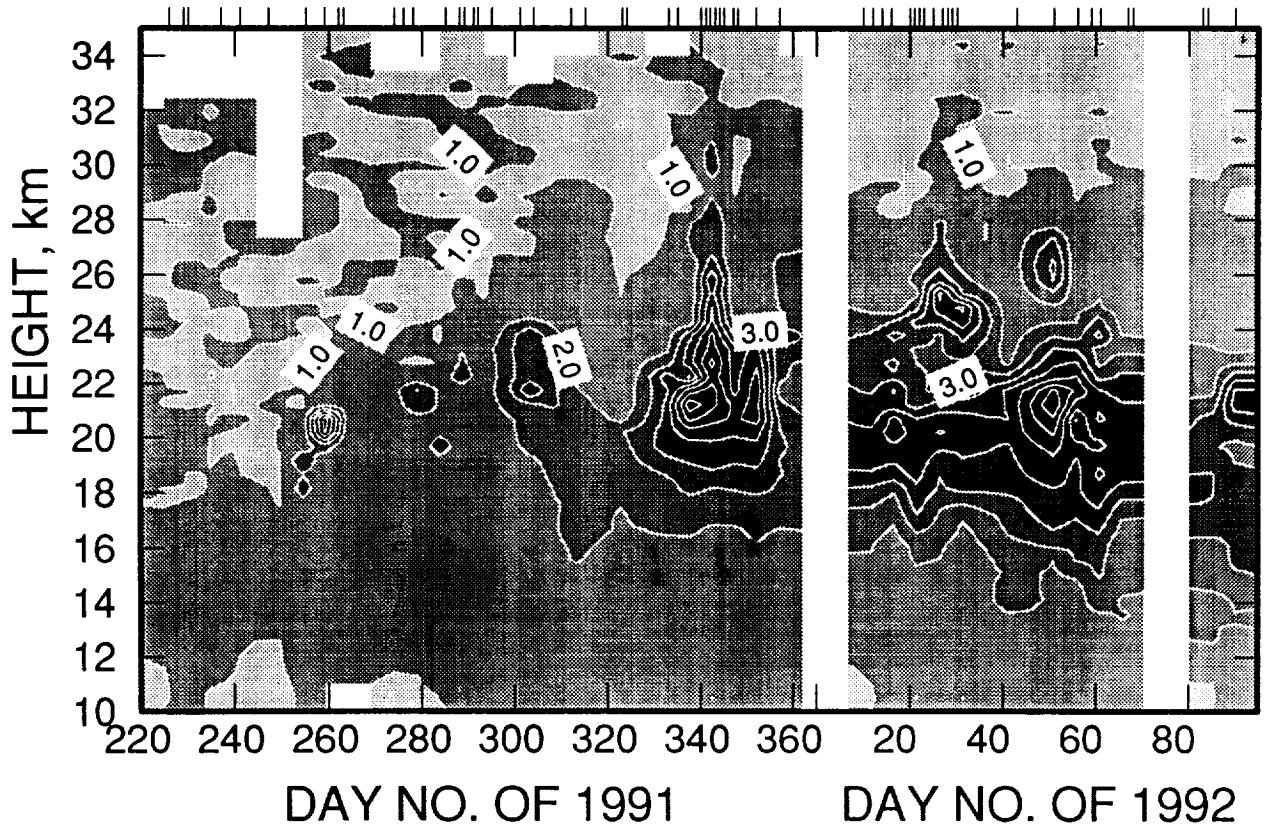

Fig.1: Lidar backscatter ratio measured by the lidar at Aberystwyth from August 1991 through March 1992

part of the European Arctic Stratospheric Ozone Experiment (EASOE). Zenith measurements only were made from 2 November to 30 April 1992, when the spectrometer wasreturned to Aberystwyth.

During this period the main bulk of the Mt. Pinatubo aerosol cloud reached the U.K. Lidar measurements at Aberystwyth monitored the cloud from August onwards: a contour plot of backscatter ratio (uncorrected for aerosol absorption) is shown in fig.1. Balloon-borne measurements made during EASOE showed that the NO_2 layer resided above the aerosol (indeed, the aerosol probably accelerated the conversion of NO_x to nitric acid), so that the effect of aerosol on the derivation of NO_2 vertical column totals is small. For ozone, on the other hand, the problem is more serious: most of the ozone lies in the height region of the aerosol layer, and the effect of aerosol scattering on the derivation of vertical column is especially pronounced for zenith angles > 90°

A preliminary correction for aerosol scattering, derived by CNRS from a balloon-borne SAOZ flight in November and from a radiative transfer model, has been applied to the data. Since the lidar data show that the amount of aerosol was very variable and increased dramatically in the early part of December caution is needed before drawing firm conclusions from these data.

4. RESULTS

a)ozone

Ozone column totals (N_v) for March-October 1991 at Aberystwyth and November 1991 - March 1992 at Lerwick are presented in figs. 2 and 3. The seasonal cycle of ozone is well

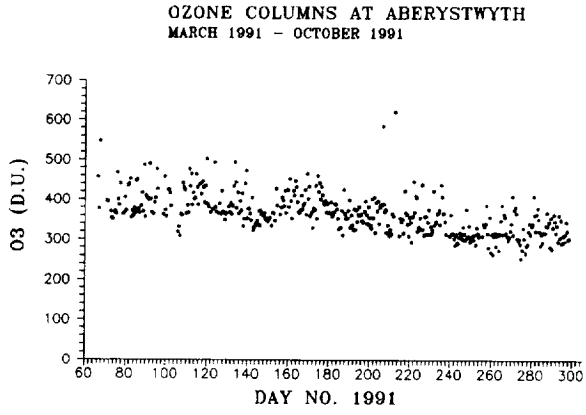

Fig.2

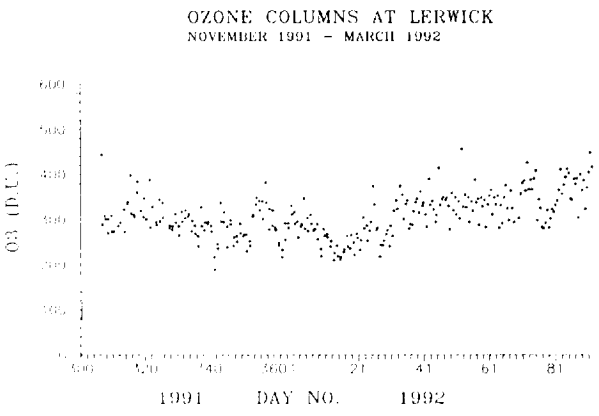

Fig. 3

represented, but it is noiceable that values in Lerwick at the end of March 1992 were about 70 DU lower than at Aberystwyth the previous year. Some of this difference may be due to the influence of aerosols, but some is likely to be real. The considerable spread in total ozone values reflects genuine day-to-day differences due to meteorological influences as well as measurement uncertainties. Error bars are not shown; the measurement precision in most cases was about 15 DU, but other sources of error (such as uncertainty in the vertical profile of ozone) can be considerably larger. On occasions, multiple scattering in the troposphere (on overcast days) can lead to values which are clearly erroneous (e.g near days 210-215, fig.2).

To gain a better understanding of the accuracy of the SAOZ, the measurements have been compared to TOMS and to the Dobson spectrophotometer at Lerwick for the period that the SAOZ spent there. The comparison with the Dobson instrument is shown in figs. 4 and 5. The agreement is generally good, except in February 1992 when the Dobson

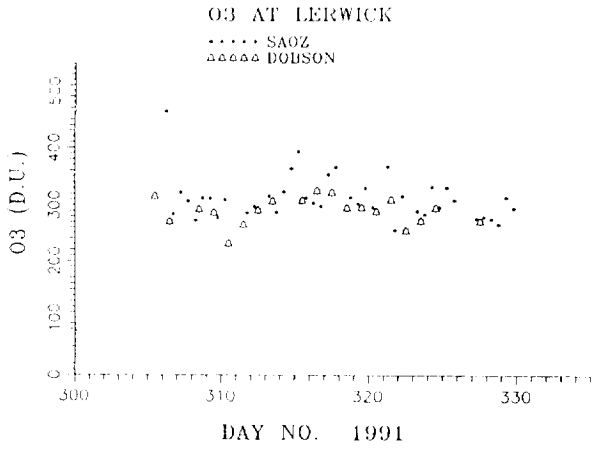

Fig. 4

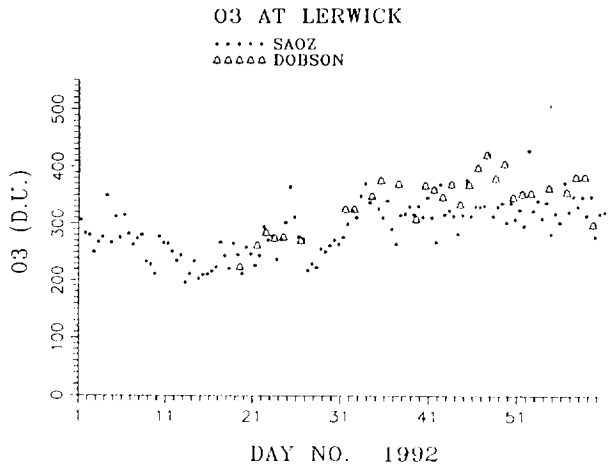

Fig. 5

exceeds the SAOZ by about 40 DU. A more consistent discrepancy is seen with TOMS (fig.6). The values shown are version 6 processed data (provided by Dr.A. Lapworth, Met. Research Unit, Cardington). The SAOZ appears to measure about 30-40 DU more than TOMS, especially in November, December and January when the agreement with Dobson is good. Consistent with this, the discrepancy between TOMS and SAOZ decreases in February, but is still evident. The most likely reason for the discrepancies, as noted above, is the influence of Mt. Pinatubo aerosol.

b)NO_2

The record of NO_2 column totals is shown in figs. 7 and 8. They demonstrate the very clear diurnal variation in NO_2 in the summer, with dusk values considerably higher than those at dawn because of photolysis of the N_2O_5 reservoir during the day. Occasional very high values are due to tropospheric pollution: the site is generally a clean one enjoying prevailing westerly winds, but when an anticyclone is established

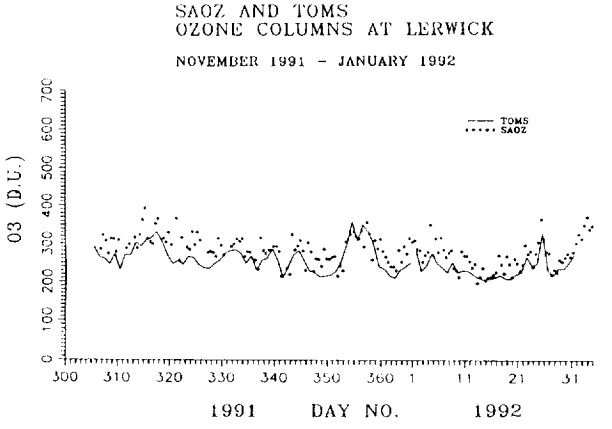

Fig. 6

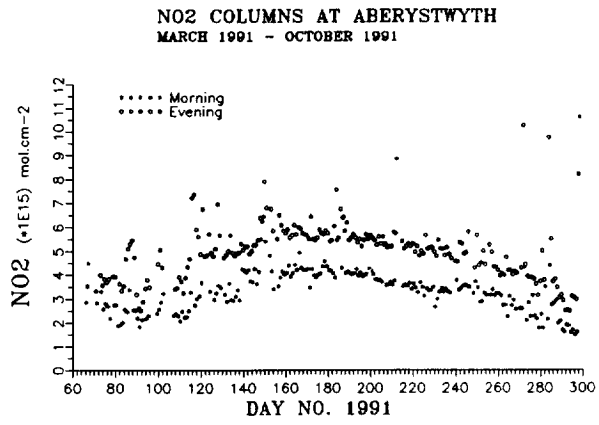

Fig. 7

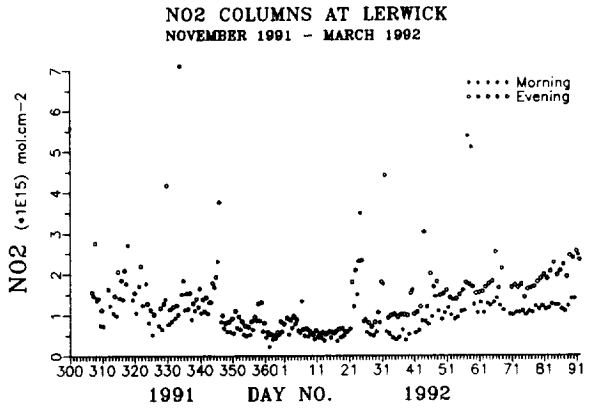

Fig. 8

near the UK polluted air is advected over it. In winter at Lerwick, NO_2 levels fell to about $1x10^{15}$ cm^{-2}, with little diurnal variation; thus, the N_2O_5 was not functioning as a reservoir species at this time, possibly due to heterogeneous conversion to HNO_3 on aerosols and (on occasion) PSCs. NO_2 levels recovered in March but were still about half of what was observed at Aberystwyth the previous year.

5. CONCLUSIONS

SAOZ measurements at Aberystwyth and Lerwick have shown that ozone and NO_2 levels over the UK in the spring of 1992 were considerably smaller than a year previously. Aerosols from Mt. Pinatubo may have affected the ozone measurements, but their influence on NO_2 is minimal and the result is real. Further investigation is required to correct the ozone column totals and to explain why the NO_2 totals were so low in 1992.

ACKNOWLEDGEMENTS

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